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Near-Net-Shape Forming of Ceramic Refractory Composite High Temperature Cartridges by VPS

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Abstract

Near-net-shape forming of high temperature furnace containment cartridges is being developed using the Vacuum Plasma Spray (VPS) process. The cartridges are thin walled, 0.069 mm (0.027 in.) thick, and have been produced in continuous lengths of 58.4 cm (23 in.). VPS has been used to deposit a refractory metal wall structure (i.e., tungsten) and coat the structure both inside and out with a ceramic (i.e., alumina). The ceramic-refractory-ceramic composite provides environmental protection to the refractory metal structure from both chemical attack (inside) and oxidation (outside). Microstructures have been characterized, and limited material properties will be presented.

THE UNITED STATES MICROGRAVITY LABORATORY onboard the Space Shuttle is dedicated to materials research in microgravity environments. The Crystal Growth Furnace (CGF), which is a part of this laboratory, is a directional solidification furnace used to evaluate the effects of microgravity on the solidification of electronic and electro-optic materials. The CGF allows on-orbit automatic and normal sample exchange of some highly toxic materials, and processes materials at temperatures up to1400°C (2552°F). The CGF made its debut on the the USML-1 Spacelab mission in 1992[1,2]. Continuance of this microgravity research will take place on USML-2 scheduled for flight in1995.

Materials processing in space offers many advantages over earth-based processing. The earth's gravitational field causes several phenomenon to occur. These are: *separation of different density* materials, introduction of convective flows in heated liquids and deformation of fragile materials under their own weight. Near zero gravity materials processing can produce materials that are very near their theoretical strength due to the elimination of these phenomenon, and the clean space environment.

One material that is of interest to microgravity researchers is gallium arsenide (GaAs). Interest in this material as a semiconductor is based on its ability to operate eight times faster than the current silicon chips and only consume one-tenth the power[3]. This material, however, is very difficult to process since it requires a very high temperature (1260°C, 2300°F) and is very corrosive in the liquid state. At these temperatures, the liquid semiconductor attacks most structural materials used to contain it during the experiments. Pyrolytic boron nitride, alumina and some refractroy metals do, however, resist chemical reaction with GaAs at these processing temperatures.

Efforts to develop and fabricate a CGF containment cartridge out of ceramics, refractory metals and combinations of ceramics and refractory metals have had limited success. The CGF containment cartridge must be structurally sound at processing temperatures and resist chemical attack by GaAs. The cartridge is of a test tube configuration with an outside diameter of 2.5 cm (1 in.), a wall thickness of 0.069 mm (0.027 in.) and a length of 58.4 cm (23 in.). The limited success of the previous efforts led to the investigation of Vacuum Plasma Spray Forming as a method to near-net-shape form a CGF cartridge[3]. VPS was identified as a result of its unique ability to spray a wide variety of metals, ceramics and metal-ceramic combinations[5,6].

Experimental Procedure

Powder - Development has concentrated on tungsten and alumina based on their relatively low costs compared to other refractory materials, but more importantly on preliminary compatibility testing[7,8,9]. Other materials, such as rhenium, molybdenum and tungsten-rhenium, have also been sprayed and evaluated. This testing has indicated that these materials would likely be suitable for a GaAs containment cartridge. Each of the powder lots received were evaluated for size distribution via a Microtrac™ and morphology via scanning electron microscopy. All of the powders were found to be very angular; none were gas atomized. The powders were typically -325 mesh.

Spray Mandrels - Leachable or removable mandrel development efforts were reported previously[4]. It was determined that graphite would survive the harsh, high temperature VPS environment. Since that time, however, it was found that several grades of graphite worked extremely well for spray forming tungsten. The mandrels were fabricated to the exact inside contour of the containment cartridge.

VPS Parameter Development - Optical microscopy was used to evaluate the microstructural characteristics of each deposit. Deposits were qualitatively evaluated for characteristics such as density, melted and unmelted particles, and grain size/shape. This information was fed back to allow appropriate adjustment to spray parameters and gun manipulation computer programs.

Spray Procedure Development - This work was conducted in the NASA-MSFC VPS Laboratory. The lab is equipped with a standard DC plasma spray system manufactured by Electro-Plasma Inc., which has been modified extensively with advanced vacuum robotics and controls to spray complex parts. A

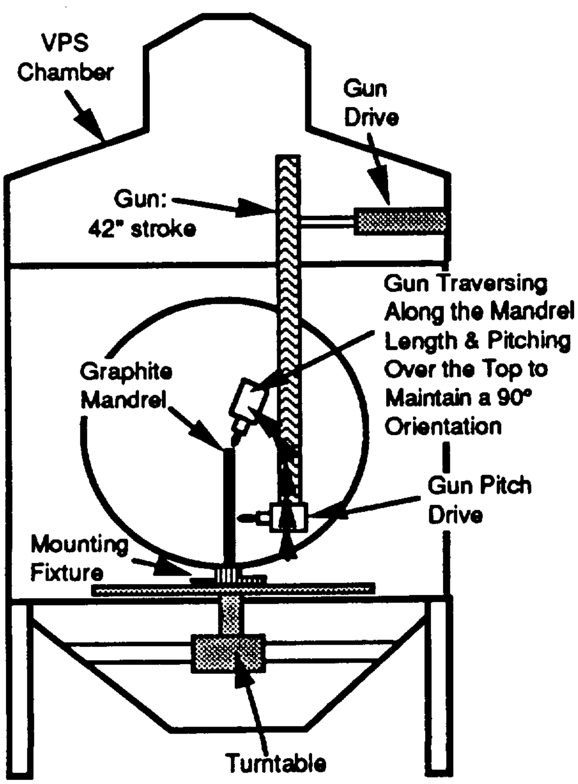


Fig. 1 - VPS Spray Configuration in the NASA-MSFC Chamber

schemetic of the system set-up for spraying the containment cartridges is shown in Figure 1.

Several spray techniques were investigated to produce high integrity deposits. Of particular interest was the ability to produce a containment cartridge that would not leak under 15 psi helium and had a uniform coating thickness circumferentially as well as along the cartridge length. The turntable rotational speed and hence, the mandrel surface speed, was held constant at 40 RPM. An Allen Bradley 8200-AT CNC programmable controller was used to control vacuum robotics inside the chamber to manipulate the plasma gun along the length of the mandrel, maintaining the gun at 90° to the surface at all times.

Testing - Limited GaAs compatibility testing was conducted on VPS tungsten, alumina and tungsten-rhenium. This work was performed for NASA-MSFC by The University of Alabama at Birmingham. Here the material of interest is sealed in a pyrolytic boron nitride crucible with GaAs and exposed to 1260°C (2300°F) for 70 hours. The samples are then examined via optical and scanning electron microscopy to determine whether a reaction occurred.

A limited number of compression tests of 2.5 cm (1 in.) tube segments were also conducted. The tube segments were laid on their side and compressed until failure. This was performed to determine the burst pressure of the VPS tubes.

Leak testing was conducted on each VPSed spray formed containment cartridge. The tubes were pressurized with 15 psi helium and submersed into a water bath. The tubes were then visually examined for leaks indicated by bubbling.

Results & Discussion

Powder - During the spraying development efforts several different lots of tungsten powder were received from the same vendor. It was found that certain lots created a lot of dust in the chamber during and after spraying and resulted in a lower quality deposit. The dusting problem was attributed to fines present in certain powder lots. Apparently, according to the vendor, there were several different methods of preparing tungsten powder and the powder precursor is very important to eliminate the fines and therefore, the dusting problem.

Spray Mandrels - Graphite that had a low volume percent binder was used to eliminate excessive outgassing or volatilization during the spray run. The VPS deposits could be removed after the spray runs because of the thermal expansion difference. These mandrels were also reusable up to 3-4 times. At that point the mandrel starts to pit and decrease in size (O.D.) due to a reaction with the tungsten to form tungsten carbide.

VPS Parameter Development - Tungsten spray development was conducted by depositing onto stainless steel plates and evaluating the

Table 1. Tungsten Spray Parameters

Run	Arc Gas Flowrate H_2:Ar (SLM)	Volts	Amps	Powder Gas Flowrate, SLM	Vac., torr
1	24.5; 78.4	61	1600	6.6	190
2	24.5; 92.0	62.7	1600	6.1	195
3	29.3; 82.6	63.5	1600	5.7	175
4	31.6; 102.4	65.5	1600	5.7	180
5	31.6; 93.0	65	1600	5.7	140
6	31.6; 93.0	65	1600	5.7	100

microstructures with optical microscopy. The deposits were qualitatively analyzed for tungsten grain characteristics (splat vs. recrystallized), density and unmelted powder particles. The parameters investigated are listed in Table 1. Rapid feedback from microstructural analysis allowed appropriate changes to be made to spray parameters prior to the next run.

Parameters from Run #6 were found to produce the best microstructural results. A photomicrograph of this deposit is shown in Figure 2. A tungsten-25%rhenium alloy powder blend was also sprayed using the parameters from Run #6. The microstructure of this deposit is shown in Figure 3. It consists of a high-density, layered structure with isolated W and Re islands.

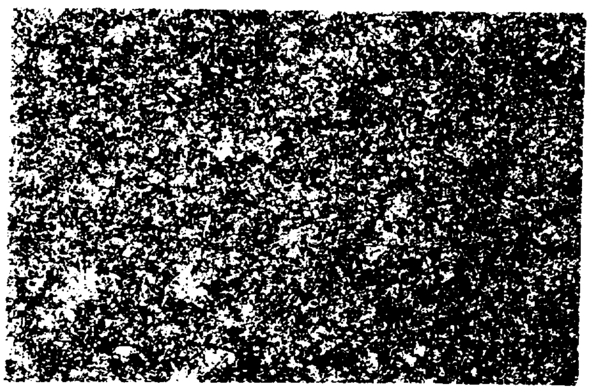


Fig. 2 - Vacuum plasma tungsten sprayed using optimized parameters. 100x (Etchant - Modified Murakami's Reagent)

Spray Procedure Development - Early efforts concentrated on what was termed a "repeated traverse" spray. This consisted of traversing the gun along the length and pitching over the tip of the mandrel at a speed of 1143 cm/min (450 in/min) with the turntable rotating at 40 rpm. The gun was used as the primary heat source. Previous work showed that a substrate spray temperature of over 1093°C (2000°F) must be maintained to insure a high integrity deposit[4]. With this method it was difficult to maintain

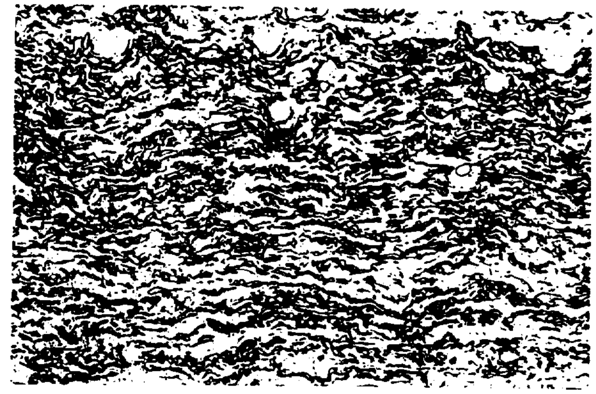
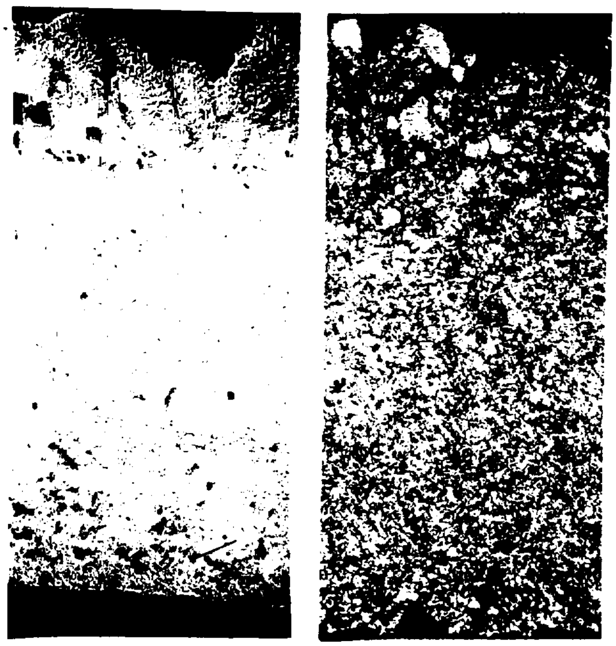


Fig. 3 - Vacuum plasma sprayed tungsten-25%rhenium blend. 100x (Etchant - Modified Murakami's Reagent)

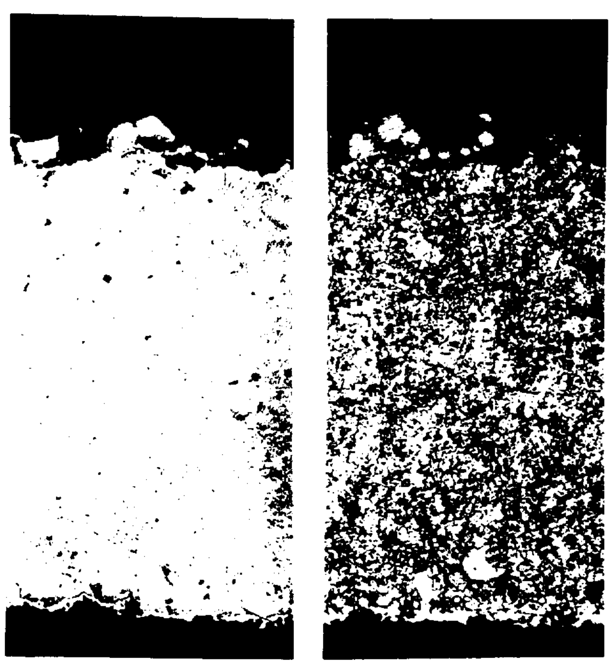
temperatures over 1093°C (2000°F) along the entire 61 cm (24 in.) length of the mandrel. In addition, this technique did not produce a uniform coating thickness, which was a consequence of the spiral application of the coating onto the mandrel. Differences in coating thickness were as much as 0.25 mm (0.010 in.) and 0.51 mm (0.020 in.) around the circumference and along the length, respectively.

These difficulties led to the investigation of an alternate method termed "creep" spray. This consisted of a single traverse of the gun over the mandrels. Speeds of 25.4-35.6 cm/min (10-14 in/min) over the mandrel tip, due to the concentrated heating as the gun pitches over the tip, and 2.5-5.1 cm/min (1-2 in/min) along the mandrel body were used. Temperatures over 1316°C (2400°F) were maintained throughout the runs. Further, differences in coating thickness were greatly reduced. Circumferential and longitudinal thickness variations were reduced to less than 0.025 mm (0.001 in.) and 0.127 mm (0.005 in.), respectively. The one problem with the method, however, was the leading and trailing overspray fringes of the plasma flame which tended to deposit somewhat porous layers on the I.D. and O.D. of the deposit with a very dense, recrystallized middle section. A photomicrograph of this phenomenon is shown in Figure 4. In the hopes of eliminating these porous layers, reverse transfer arc was incorporated into this spray technique. A slight improvement was noticed and is shown in Figure 5 for comparison purposes.



A) Unetched B) Etched

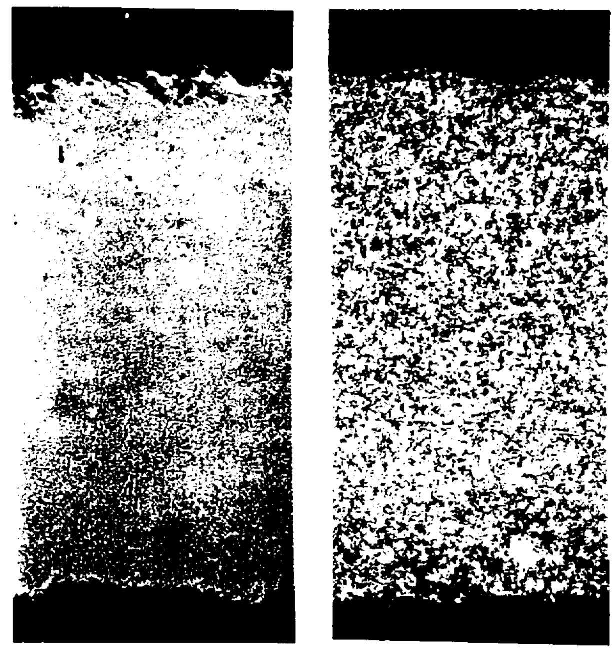
Fig. 4 - Photomicrograph of vacuum plasma sprayed tungsten showing the leading and trailing overspray phenomenon associated with the creep spray technique. A) Unetched, B) Etched, 100x (Etchant - Modified Murakami's Reagent)



A) Unetched B) Etched

Fig. 5 - Vacuum plasma sprayed tungsten using the creep spray technique with reverse transfer arc. A) Unetched, B) Etched, 100x (Etchant - Modified Murakami's Reagent)

To further address healing the porosity, a postheat using the plasma gun as the heating source was introduced. It was performed without the powder on and at the creep spray speeds discussed above. This also showed improvement and improvement in the tungsten microstructure as witnessed in Figure 6.



A) Unetched B) Etched

Fig. 6 - Vacuum plasma sprayed tungsten using the creep spray technique and a postheat. A) Unetched, B) Etched, 50x (Etchant - Modified Murakami's Reagent)

Testing - GaAs was reported to have penetrated the surface of VPS samples of tungsten, alumina and tungsten-25%rhenium. It did not, however, react with these materials. This was due, apparently, to surface porosity associated with these deposits.

A tungsten tube section tested in compression failed at a load of 19 pounds. A stress analysis performed, using this failure load, indicated this would result in a shell failure stress of 57.4 ksi.

Approximately 80% of the tubes produced by the creep spray method passed leak testing when pressurized with 15 psi helium. This percentage was improved to 90% with the incorporation of the reverse transfer arc and postheat techniques.

Conclusions

1. Tungsten can be near-net-shape spray formed.
2. Graphite mandrels survived the harsh VPS environment and were reusable up to 4 times.
3. The creep spray method produced more uniform coating thicknesses due to its slow traverse speed.

4. The creep spray method maintained the part spray temperature above 1316°C (2400°F).
5. Reverse transfer arc and postheating improved deposits sprayed using the creep spray technique.
6. VPS tungsten, alumina and tungsten-rhenium have shown resistance to high temperature attack by GaAs.

Acknowledgments

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